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ELECTRON BEAM PUMPING OF LASER DYES

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We report a study into the feasibility of electron beam pumped dye lasers. This work was undertaken because such lasers might be capable of producing powerful (e.g., 10^6 - 10^9 watts) nanosecond pulse of visible light with a conversion efficiency as high as 5% from electrical energy into light. However, laser action could not be attained. We propose that high energy radiation results in indiscriminate ionization and excitation of the supporting solvent for the dye and hence a transient absorption of the fluorescent light.

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ELECTRON BEAM PUMPING OF LASER DYES

by

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FINAL TECHNICAL REPORT

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We report a study into the feasibility of electron beam pumped dye lasers. This work was undertaken because such lasers might be capable of producing powerful (e.g., 10^6 - 10^9 watts) nanosecond pulse of visible light with a conversion efficiency as high as 5% from electrical energy into light. However, laser action could not be attained. We propose that high energy radiation results in indiscriminate ionization and excitation of the supporting solvent for the dye and hence a transient absorption of the fluorescent light.

Experimental

The electron beam was produced by a Febetron 706 accelerator operated with at a mean electron energy of 0.5 meV and a maximum current density of 5×10^3 amp cm^{-2}. The input pumping power could be attenuated over a range of $\sim$ 100 by suitable spatial filtering of the beam. In this way, it is possible to maintain the same electron kinetic energy and thus penetration, while varying the pumping power. Spectrographic measurements were made on a "small" Hilger quartz prism spectrograph, covering 185 nm - 700 nm on a single plate. Time-resolved measurements were made with a calibrated S-20 biplanar photodiode, in conjunction with Tektronix 519 and 7904 oscilloscopes. The dyes were excited in a transverse geometry with a high Q cavity (99.9% and 95% reflectivity mirrors) and solutes were highly purified and could be deoxygenated if necessary.

Results and Discussion

As there have not been any previous studies of electron beam

excitation of laser dyes at these high intensities, we first undertook to study representation dye/solvent systems for a range of laser dyes to find the most strongly emitting combinations. The correct choice of solvent is essential, since excitation must be transferred to the dye following bulk solvent excitation. Following this initial screening to narrow the selection, a comprehensive study over a range of experimental parameters such as pumping power, dye concentration, and added quenchers was undertaken. The results are as follows:

a. Polar solvents - water, alcohols

The majority of widely used laser dyes are soluble in polar solvents. These are, for example, xanthene, coumarin, and polymethine dyes. It is, however, well known (1) that excited states of water and alcohols are all autoionizing Ryderg states and there is no energy transfer to the solute. The possibility remained, however, of utilizing chemiluminescent formation of excited states of dye molecules as follows (2),

$$e^-_{aq} + \text{dye} \rightarrow \text{dye}^-$$
$$\text{dye}^- + \text{OH} \rightarrow \text{dye}^* \rightarrow \text{dye} + h\nu.$$

We studied the light emission from Rhodamine 6G, ariflavine, sodium fluorescein, and 7-diethyl-4-methyl coumarin in aqueous and alcoholic solution. In all cases, the spontaneous emission was extremely weak, $\sim 10^{-3}$ of the predicted 0.01% efficiency estimated using known radiolytic yields. This very low luminescence efficiency is probably attributable to the intrinsic concentration of the transient dye anion. These systems were not further pursued.

b. The highly polar dyes described above will dissolve to a degree in weakly polar solvents such as 1,4-dioxane. This solvent has the advantage that it does transfer electronic excitation to solute molecules, albeit with an efficiency of $\sim$ 0.3%. The well-documented dye 7-diethyl-4-ammocoumarin proved to be particularly soluble, although because of the low polarity of the solvent it formed dimers which do not fluoresce as well due to enhanced intersystem crossing rates. In this case, at low concentrations and dose, the luminescence efficiency approached 0.1%, but at high concentrations and excitation density the efficiency dropped to 0.01%. Transient absorptions were again the source of this lowered efficiency. Laser action could not be achieved because of these intrinsic intracavity losses.

c. A number of "scintillators" have been used as laser dyes (3), with fast rise-time pumping. Furthermore, these compounds have the highest energy efficiency for luminescence, approaching 5% for aromatic solvents. The p-terphenyl/cyclohexane and 9,10-diphenylanthracene/p-xylene systems serve to illustrate the results. In aromatic solvents, the energy efficiency for luminescence is highest ($\sim$ 5%), but solvent absorptions (from radicals and triplet states) restrict the useful spectral region to 425-525 nm. With these constraints, the best scintillator in this range is 9,10-diphenylanthracene, which lases at 432.6 laser pumped (4). At low concentrations and excitation density (the efficiency is a function of concentration) theoretical efficiencies are achieved, but at the higher concentrations and pumping powers required for laser action,

there is a net loss of ≥ 100% per pass over the entire fluorescent band. We suppose that this loss arises from triplet states of both solute and solvent, and could not be suppressed by the addition of cis-butene to quench these states. There is no evidence for triplet-triplet annihilation effects.

The aliphatic hydrocarbons have a much lower triplet yield than aromatic compounds. In this case the most favorable combination is p-terphenyl in methylcyclohexane. In fact, p-terphenyl has a significant advantage because its triplet absorption (5) does not overlap the fluorescence. Furthermore, the aliphatic hydrocarbons are completely transparent (6) in the near u.v. (346 nm) where p-terphenyl fluoresces. In this case, the energy efficiency approached 1% at the high concentrations and excitation density necessary for laser action. Measurement of the gain and loss in the laser cavity, however, showed that there was still sufficient loss to prevent laser action even when 20 times over the threshold observed for optical pumping (7). This loss is smaller than observed in any of the other systems previously described, being ≥ 80% per pass. The absorbing species here is the p-terphenyl cation, which is the precursor of the fluorescing state. Although this species should have a very short lifetime at high scintillator concentrations, it was not possible to completely quench it away by increasing the concentration of either the scintillator or positive charge scavengers.

Conclusions

The results of this study are that radiation-induced transient species, produced concomitantly with the desired fluorescing state, absorb sufficiently to prevent laser action when attempting to pump dye lasers with electron beams. Successful electron beam excitation of dye lasers is not impeded by any fundamental physical effect, rather by the sensitivity of present dye laser systems to the indiscriminate excitation processes intrinsic to ionizing radiation. We intend to continue working on finding a suitable dye/solvent combination.

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Appendix

During the time of this contract we have also been active in electron beam excitation of vacuum ultraviolet lasers in high pressure noble gases. We have succeeded in achieving laser action in xenon using the Febetron 706 accelerator, with substantially lower pump energy (~ 6 J vs. 400 J for other work) by carefully optimizing excitation efficiency. This work is detailed in the two papers attached to this report.